

Onset of metallic ferromagnetism in a doped spin-orbital chain

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Starting from a spin-orbital model for doped manganites, we investigate a competition between ferromagnetic and antiferromagnetic order in a one-dimensional model at finite temperature. The magnetic and orbital order at half filling support each other and depend on a small antiferromagnetic superexchange between t_{2g} spins and on an alternating Jahn-Teller potential. The crossover to a metallic ferromagnetic phase found at finite doping is partly suppressed by the Jahn-Teller potential which may localize e_g electrons. [published in *phys. stat. sol.* **242**, 311 (2005)]

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1 Introduction The transition from an insulating to metallic behavior in doped manganites, observed as a function of temperature or filling, is one of the outstanding problems in the field of strongly correlated systems. The theoretical challenge is to understand a rich variety of metallic, insulating, magnetically ordered and orbital ordered phases of doped manganites in terms of the dynamics of correlated e_g electrons which involves their orbital degrees of freedom [1]. Magnetic interactions: spin superexchange (SE) and double exchange (DE), as well as orbital SE, frustrate each other and their competition leads to several magnetic phases, including phase separation [2].

While orbital order supports *A*-type antiferromagnetic (AF) phase in LaMnO_3 [3], one expects that e_g orbitals play an important role in the insulating ferromagnetic (FM) phase at finite doping, below the crossover to a metallic FM phase [1]. Here we investigate a one-dimensional (1D) model derived from the realistic spin-orbital model for manganites [3], showing an interplay between spin and orbital order.

2 Effective orbital t - J model at finite temperature We investigate the 1D orbital t - J model for manganites for a given configuration of core spins \mathcal{S} ,

$$\mathcal{H}(\mathcal{S}) = H_t + H_J + H_{J'} + H_{JT}, \quad (1)$$

which consists of the hopping term H_t , the orbital superexchange H_J for the e_g electrons, superexchange $H_{J'}$ for the core spins formed by t_{2g} electrons, and an alternating Jahn-Teller (JT) potential H_{JT} .

As we consider 1D chains in z -direction, only electrons within $|z\rangle \equiv \frac{1}{\sqrt{6}}|3z^2 - r^2\rangle$ orbitals are mobile, while those within $|x\rangle \equiv \frac{1}{\sqrt{2}}|x^2 - y^2\rangle$ orbitals are localized:

$$H_t = - \sum_i t_{i,i+1} (\tilde{c}_{iz}^\dagger \tilde{c}_{i+1,z} + \tilde{c}_{i+1,z}^\dagger \tilde{c}_{iz}), \quad (2)$$

where \tilde{c}_{iz} and \tilde{c}_{iz}^\dagger are the annihilation and creation operators restricted to the Hilbert space without double occupancies. Due to strong Hund's coupling between e_g and t_{2g} electrons, the hopping amplitude, $t_{i,i+1} =$

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$tu_{i,i+1} = t \cos(\theta_{i,i+1}/2) e^{i\chi_{i,i+1}}$, depends of the relative angle $\theta_{i,i+1}$ of the two classical core spins at sites i and $i+1$, see e.g. Ref. [4]. The complex phase $\chi_{i,i+1}$ can be neglected for a 1D chain [5].

It is convenient to introduce orbital pseudospin operators $T_i^z = \frac{1}{2}\sigma_i^z = \frac{1}{2}(n_{ix} - n_{iz})$, with eigenstates $|z\rangle$ and $|x\rangle$. With the electron number operator \tilde{n}_i restricted to the Hilbert space without double occupancies, the superexchange term which follows from Refs. [3] and [6] becomes

$$H_J = \frac{1}{5}J \sum_i (2|u|_{i,i+1}^2 + 3) \left(2T_i^z T_{i+1}^z - \frac{1}{2}\tilde{n}_i \tilde{n}_{i+1} \right) - \frac{9}{10}J \sum_i (1 - |u|_{i,i+1}^2) \tilde{n}_{iz} \tilde{n}_{i+1,z} \\ - J \sum_i (1 - |u|_{i,i+1}^2) [\tilde{n}_{iz}(1 - \tilde{n}_{i+1}) + (1 - \tilde{n}_i)\tilde{n}_{i+1,z}]. \quad (3)$$

Like the kinetic energy, it depends on the direction of core spins via the factors $|u|_{i,i+1}^2$. The first term $\propto (2|u|_{i,i+1}^2 + 3)$ favors FM spin order for ion pairs $\text{Mn}^{3+}-\text{Mn}^{3+}$, while the second and third ones $\propto (1 - |u|_{i,i+1}^2)$ favor AF order for $\text{Mn}^{3+}-\text{Mn}^{3+}$ pairs [$\propto \tilde{n}_{iz}\tilde{n}_{i+1,z}$], and for $\text{Mn}^{3+}-\text{Mn}^{4+}$ pairs [$\propto \tilde{n}_{iz}(1 - \tilde{n}_{i+1})$]. The SE parameter $J = t^2/\varepsilon(^6A_1)$ is given by the high-spin excitation energy $\varepsilon(^6A_1)$ and the prefactors of the above terms were chosen to preserve the relative magnitude of the different excitations for realistic parameter values [3]. Throughout this work, we use $J = 0.125t$ and choose t as unit of energy.

The third term in Eq. (1) is an AF Heisenberg interaction between the core t_{2g} spins,

$$H_{J'} = J' \sum_i (\vec{S}_i \cdot \vec{S}_{i+1} - S^2). \quad (4)$$

As we used classical core spins \vec{S}_i of unit length, their physical value $S = 3/2$ was compensated by a proper increase of J' . Finally, we include an alternating JT potential of the form

$$H_{\text{JT}} = 2E_{\text{JT}} \sum_i \exp(i\pi R_i) T_i^z, \quad (5)$$

We employ a Markov chain Monte Carlo (MCMC) algorithm for the classical core spins (see e.g. Ref. [4]) combined with exact diagonalization for the electron degrees of freedom [7]. In this method, the many-particle Hamiltonian determined by $\{u_{i,i+1}\}$, which are in turn given by the core spin configuration \mathcal{S} , is solved by exact diagonalization taking into account its block-diagonal structure. The lowest states of each block are used to evaluate the trace over the fermionic degrees of freedom, $\text{Tr}_c e^{-\beta \mathcal{H}(\mathcal{S})} =: w(\mathcal{S})$, which gives the statistical weight for \mathcal{S} and which is sampled by the MCMC. Autocorrelation analysis was used in order to ensure that enough configurations were skipped between measurements.

3 Numerical results and discussion We begin with analyzing spin and orbital correlations at increasing doping. Figure 1 shows the core spin structure factor, $S(k) = \sum_{i,j} S_i S_j e^{-ik(i-j)}$, for an $N = 12$ chain and doping $0 \leq x \leq 5/12$ ($x = 1 - n$, where n is electron filling) near the system filled by $n = 1$ electron per site (as in LaMnO_3), and for three different parameter sets at low temperature. The orbital structure factor, $T(k) = \sum_{i,j} T_i^z T_j^z e^{-ik(i-j)}/n^2$, which is renormalized by the squared electron density for better comparability of results obtained at different fillings, for the same parameters is shown in Fig. 2. Altogether, the orbital and magnetic order obtained at $x = 0$ and $E_{\text{JT}} = 0$ complement each other — polarized (ferro) $|z\rangle$ orbitals at $J' = 0.02t$ induce AF spin order [Figs. 1(a) and 2(a)], but if the occupied e_g orbitals alternate, FM spin order is supported instead [Figs. 1(b) and 2(b)]. Increasing doping induces a gradual crossover towards FM order due to the DE mechanism, being the most efficient in the metallic phase when only $|z\rangle$ orbitals are occupied. This insulator-metal transition is hindered by the JT potential which favors orbital alternation [Fig. 2(c)] and suppresses FM correlations [Fig. 1(c)].

For a finite SE between core spins $J' = 0.02t$ and in absence of JT potential ($E_{\text{JT}} = 0$), a transition from AF to FM spin order is induced by doping [Fig. 1(a)], and predominantly the mobile $|z\rangle$ orbitals

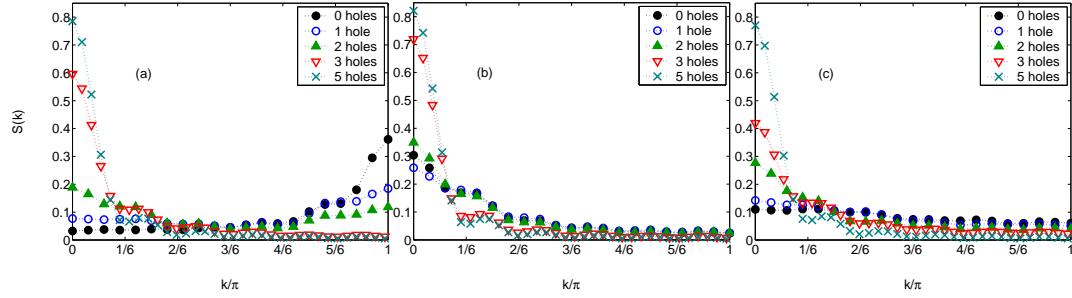


Fig. 1 Core spin structure factor $S(k)$ for: (a) $J' = 0.02t$, $E_{JT} = 0$; (b) $J' = 0$, $E_{JT} = 0$; (c) $J' = 0.02t$, $E_{JT} = 0.1t$, and several doping levels, as obtained for an $N = 12$ site chain. Parameters: $J = 0.125t$ and $\beta t = 100$.

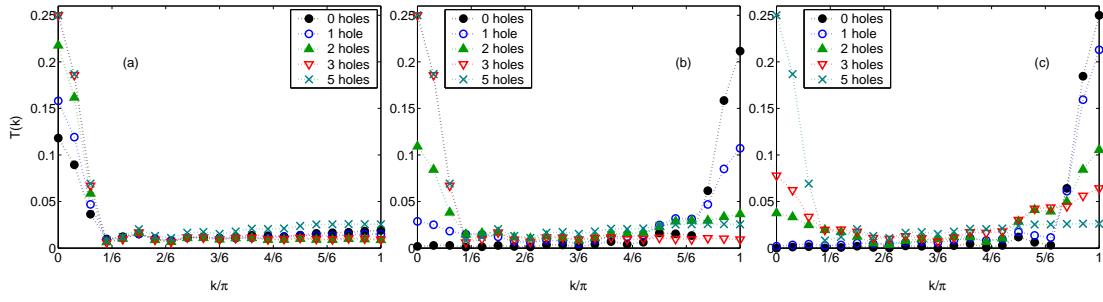


Fig. 2 Orbital structure factor $T(k)$ for: (a) $J' = 0.02t$, $E_{JT} = 0$; (b) $J' = 0$, $E_{JT} = 0$; (c) $J' = 0.02t$, $E_{JT} = 0.1t$, and several doping levels, as obtained for an $N = 12$ site chain. Parameters: $J = 0.125t$ and $\beta t = 100$.

are occupied at all doping levels [see Fig. 2(a)]. In the spectral density depicted in Fig. 3(a), one sees signals with low weight for the $|x\rangle$ orbitals and a rather broad incoherent peak due to the $|z\rangle$ excitations. The broad peak contains dispersionless states stemming from $|z\rangle$ electrons next to the few $|x\rangle$ electrons. Additionally, a dispersive band is found which comes from $|z\rangle$ electrons moving in the AF background, showing a remarkable similarity to a one-orbital model [5]. At the higher temperature $\beta t = 30$, more $|x\rangle$ orbitals are occupied ($4.53 \pm 0.07 \simeq 38\%$ vs. 2.03 ± 0.1 at $\beta t = 100$), and their weight in the spectral density increases [Fig. 3(b)]. The dispersionless broad feature becomes stronger, as the motion of $|z\rangle$ electrons is hindered by $|x\rangle$ electrons. Both effects are further enhanced when spin order weakens at very high temperature $\beta t = 10$.

A chain doped with three holes $x = 0.25$ has FM order of core spins [see Fig. 1(a)] and practically only mobile $|z\rangle$ electrons [Fig. 2(a)], due to the dominating DE mechanism which favors both FM order and ferro orbital polarization. The spectral density at low temperature $\beta t = 100$ is shown in Fig. 3(d) and one sees a coherent band with almost unrenormalized dispersion $\sim 4t$ due to e_g electrons moving in $|z\rangle$ orbitals, and only a weak signal above the Fermi energy for the immobile excitations in $|x\rangle$ orbitals. For $\beta t = 30$ [see Fig. 3(e)], the peaks of the $|z\rangle$ -band are broadened, the bandwidth is slightly reduced and a small spectral weight is induced in $|x\rangle$ states below the Fermi energy (0.12 ± 0.02 for the filling by 9 electrons). Again, these effects become more pronounced for the high temperatures $\beta t = 10$, see Fig. 3(f), where 1.18 ± 0.06 electrons occupy $|x\rangle$ orbitals. The broadening of the peaks and the reduction of the bandwidth result from core spin fluctuations weakening FM correlations, while the additional (incoherent)

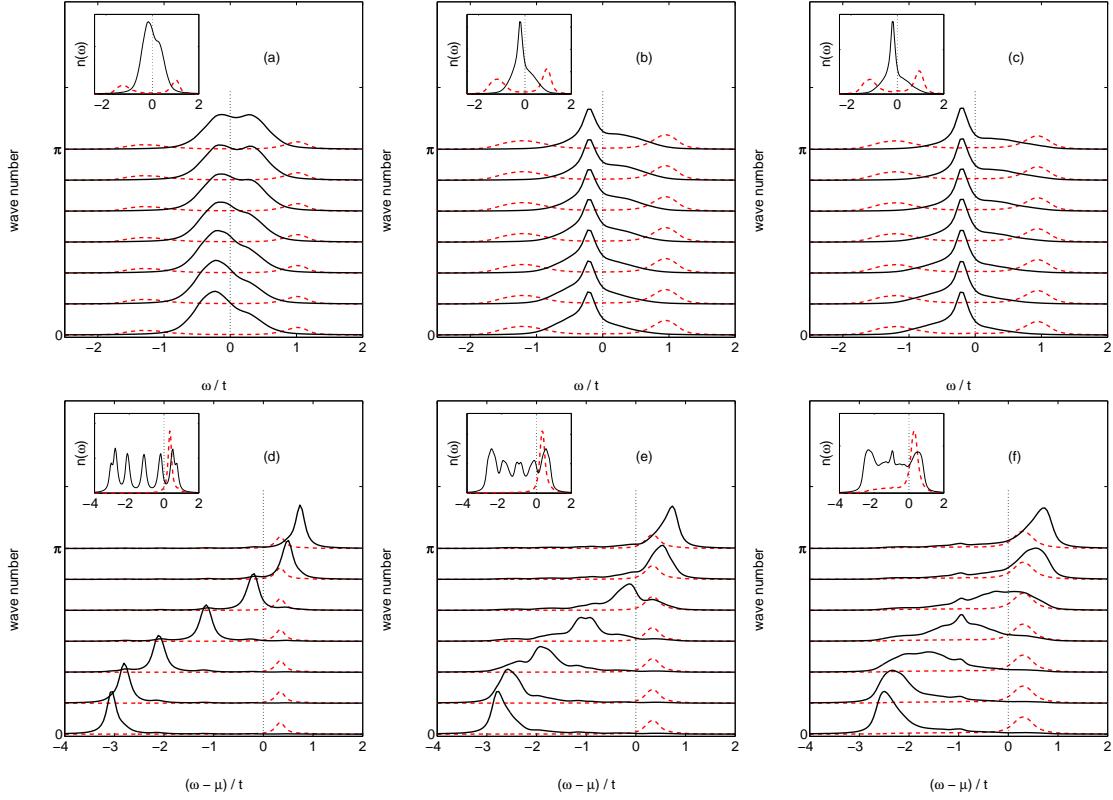


Fig. 3 Spectral functions $A(k, \omega)$ for an $N = 12$ orbital chain (1). First row — undoped chain ($x = 0$) at temperatures: (a) $\beta t = 100$, (b) $\beta t = 30$, and (c) $\beta t = 10$; second row — doped chain with three holes ($x = 0.25$) at temperatures: (d) $\beta t = 100$, (e) $\beta t = 30$, and (f) $\beta t = 10$. Solid and dashed lines for $|z\rangle$ and $|x\rangle$ excitations. Insets show the density of states $n(\omega)$. Parameters: $J = 0.125t$, $E_{JT} = 0$, $J' = 0.02t$.

signals besides the tight-binding band are caused by the $|x\rangle$ electrons. The reduction of FM correlations and the increasing electron density within $|x\rangle$ orbitals occur simultaneously with rising temperature.

Yet another situation arises (at $J' = 0.02t$) in presence of a small alternating JT potential $E_{JT} = 0.1t$. It favors alternating orbital pattern [Fig. 2(c)], which in turn destroys the core spin AF order in the undoped chain [Fig. 1(c)]. Upon doping, the initially weak FM correlations increase, $|x\rangle$ orbitals are depleted and the number of $|z\rangle$ electrons even increases slightly to ~ 7 for 2 and 3 holes. For the undoped chain there are just three peaks in the spectral function $A(k, \omega)$ [Fig. 4(a)]: a central peak from the $|z\rangle$ electrons trapped between two occupied $|x\rangle$ orbitals, and two side peaks due to the $|x\rangle$ electrons. Very similar results (not shown) are obtained for $J' = 0$ and $E_{JT} = 0$, where the orbitals also alternate, see above.

Doping with one hole removes an $|x\rangle$ electron and a three-site well is created, leading to three $|z\rangle$ signals in the spectral density depicted in Fig. 4(b). The spectral function $A(k, \omega)$ for doping with three holes [Fig. 4(c)] shows new features on top of the tight-binding-like band at $E_{JT} = 0$ (Fig. 3(d)), that arise from the two remaining $|x\rangle$ electrons and from the alternating on-site potential seen by $|z\rangle$ electrons. Note that the FM is weaker at this doping level than when the JT potential is absent, because DE is hindered. From doping $x \geq 1/3$ onward, the chain is fully polarized and FM order is enhanced, see Figs. 1(c) and 2(c). A strong JT potential $E_{JT} \sim 0.5t$ delays a crossover to the metallic FM phase to very high doping $x > 0.4$.

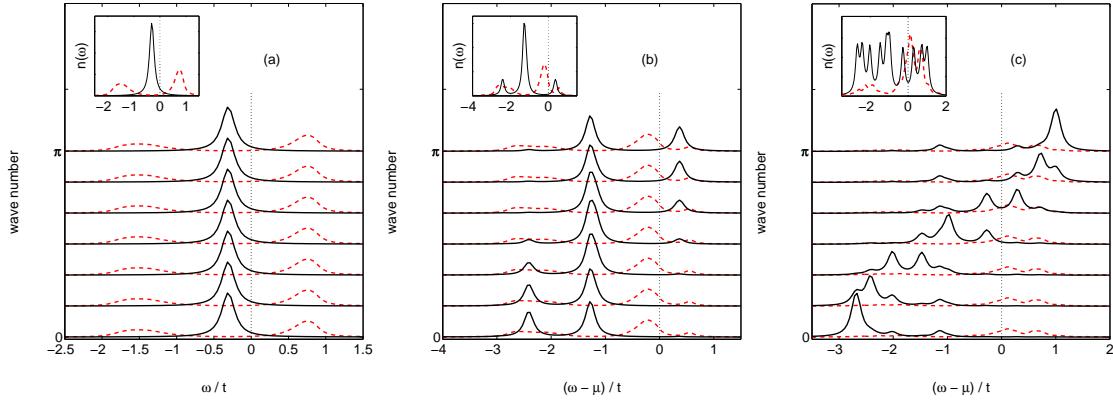


Fig. 4 Spectral functions $A(k, \omega)$ for an $N = 12$ orbital chain as in Fig. 3, but with JT potential $E_{\text{JT}} = 0.1t$, as obtained for: (a) undoped chain ($x = 0$), and chain doped with (b) one hole $x \simeq 0.083$, and (c) three holes $x = 0.25$.

Finally, we determined the DE constant for the present 1D chain, $J_{\text{DE}} = \langle H_t \rangle / (2zS^2)$, where $z = 2$ and $S = (4 - x)/2$ [6]. While the DE increases with x and is simply frustrated by the AF SE between core spins (J_{DE} does not depend on J'), increasing JT potential reduces J_{DE} in the insulating regime. For instance, at $x = 0.25$ one finds $J_{\text{DE}} \simeq 0.030, 0.027$ and $0.024t$ for $E_{\text{JT}} = 0, 0.1$ and $0.25t$. However, at doping $x = 0.42$ the FM phase is already metallic for the entire range of $0 < E_{\text{JT}} < 0.25t$, with $J_{\text{DE}} \simeq 0.046t$.

4 Summary and conclusions Even in one dimension, the orbital t - J model shows an interesting competition of FM and AF SE terms. At low temperatures and for the undoped chain, a relatively small AF SE J' between the t_{2g} core spins already suffices for a transition from FM spin order and alternating orbital order ($J' = 0$) to AF spin order and ferro orbital polarization ($J' = 0.02t$). These different situations, both encountered qualitatively in LaMnO_3 , result in quite distinct one-particle spectra. Because of the close interplay of orbital- and spin-degrees of freedom, the change between these two scenarios can also be triggered by an alternating JT potential ($E_{\text{JT}} = 0.1t$). At higher temperatures, the competing interactions lead to further changes in the spectra as both the spin and the orbital order are destroyed.

For the doped chain ($x = 0.25$) without JT potential, the kinetic energy favors FM order and orbital polarization. This metallic FM phase may be destroyed either by finite JT potential, or by increasing temperature, when both the FM order and the orbital polarization are reduced, leading in both cases to incoherent spectra with reduced bandwidth. This behavior indicates that an insulating phase may be induced either by increasing temperature, or by enhancing the JT distortions by chemical substitution. Summarizing, the present study highlights: (i) a complex interplay between spin, orbital and charge degrees of freedom, and (ii) the importance of the coupling to the lattice — all these all these aspects decide about spin and orbital correlations in doped manganites and have to be included in their realistic microscopic models[8].

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